Ordered bicontinuous double-diamond morphology in subsaturation nuclear matter

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We propose to identify the new "intermediate" morphology in subsaturation nuclear matter observed in a recent quantum molecular dynamics simulation with the ordered bicontinuous double-diamond structure known in block copolymers. We estimate its energy density by incorporating the normalized area-volume relation given in a literature into the nuclear liquid drop model. The resulting energy density is higher than the other five known morphologies.

PACS numbers: 21.65.+f, 26.50.+x, 26.60.+c

Understanding the form of existence of nuclear matter in extreme environments is of importance both from the point of view of nuclear many body problem and from the context of nuclear astrophysics. Nuclear matter with subsaturation densities ($0.1n_{\rm s}$ – $n_{\rm s}$; $n_{\rm s}$ being the saturation density) is believed to exist in the inner crust of neutron stars and to appear at stellar collapses. This density range corresponds to the transitional region between a Coulomb lattice of spherical nuclei and a uniform matter; Ravenhall et al. [1] and Hashimoto et al. [2] showed that nuclear matter experiences various phases in the course of density change. After that, this was elucidated by various model calculations [3, 4, 5, 6, 7, 8, 9]. With the help of the recent progress of computer power, not only the ground state at each density but also the dynamical phase transitions between them and the excited states were studied by means of first-principle numerical simulations with the quantum molecular dynamics (QMD) method [10, 11, 12, 13]. Among them, the most basic is to understand the form of existence of nuclear matter at each density. Watanabe et al. [11] found, in addition to the five known morphologies — sphere, cylinder, slab, cylindrical hole (tube), and spherical hole (bubble) —, a new "intermediate" morphology that is characterized by negative Euler characteristic, $\chi < 0$. They described this as a highly connected spongelike shape, and conjectured its relevance to astrophysics.

However, these five morphologies were known [14] in block copolymers of, for example, styrene and isoprene. In the nuclear case, the two domains are composed of matter and void (or very dilute neutron vapor), whereas they are composed of two kinds of polymers such as polystyrene (PS) and polyisoprene (PI) in the macromolecule case. Their morphologies are determined by a balance between the surface energy and the Coulomb energy in the former, whereas by that between the (inter-)surface energy and the stretching free energy, like that causes rubber elasticity, in the latter. A new morphology was found experimentally in a star block copolymer by Thomas et al. [15] and in a diblock copolymer by Hasegawa et al. [16] between the PS cylinder and the lamella (slab) and between the lamella and the PI cylinder. These experiment determined essentially the correct morphology (Fig. 4 in Ref. [15] and Fig. 3 in Ref. [16]). Soon thereafter the shape of the interface between the two microphases was mathematically recognized as the H surface — a family of surfaces with constant mean curvature H [17]. The H surfaces are known to minimize the area under the symmetry and volume conservation condition. The observed morphology is called the ordered bicontinuous double-diamond (OBDD) structure according to its symmetry. The OBDD structure consists of two interwoven networks of tetrahedral units (four fold junctions) filled by one material and the remaining matrix filled by the other. Calculations of its free energy were done by several groups [17, 18, 19] and they concluded that the OBDD structure is not the ground state at any PS/PI composition.

Both from the location — adjacent to the slab — of the "intermediate" phase of Watanabe et al. and their observation that it is highly connected spongelike, it looks quite natural to interpret this phase as the OBDD structure observed in block copolymers. A direct calculation of the energy density of this structure based on some microscopic nuclear Hamiltonian is desirable, but it is too much complicated unfortunately. Alternatively, here we estimate its energy density by combining the familiar liquid drop model relation and the normalized area-volume relation of the OBDD morphology given in the literature [17].

The liquid drop model relations are taken from Ravenhall et al. [1] who first predicted the non-spherical morphologies. Under the Wigner-Seitz cell approximation, the total energy density is given by $E_{\rm S}+E_{\rm C}+E_{\rm B}+E_e$, a sum of the surface energy, the Coulomb energy, the bulk energy, and the kinetic energy of electron gas. Here the Coulomb energy consists of the nuclear electrostatic energy and the lattice energy of electron gas and a spatially spread nucleus embedded in it. Zero temperature is assumed. The model is formulated by extending that for the spherical case given in Ref. [20]. We consider spherical ($d = 3$), cylindrical ($d = 2$), and slab ($d = 1$) cases as

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in Ref. [1]. The unit cell for each case is a sphere with the radius r_c, a cylinder with the radius r_c and the length l, and a rectangular parallelepiped with the sides $2r_c$, a, b, respectively. The average density over the cell is n. In each cell, nuclear matter is put in the form of a sphere with the radius r, a cylinder with the radius r and the length l, and a rectangular parallelepiped with the sides $2r$, a, b, respectively. The density of nuclear matter is n'. The other part of the cell is occupied by a very dilute neutron vapor. The proton fraction is x. The volume fraction is $u = n/n' = (r/r_c)^d$. The surface tension is σ. Among the total energy density, E_S and E_C depend on the shape and size of the cell and given by

$$E_S = \frac{u\sigma d}{r},$$
$$E_C = 2\pi n'^2 x^2 e^2 r^2 u f_d(u),$$
$$f_d(u) = \frac{1}{d+2}\Big[\frac{2}{d-2}\Big(1 - \frac{1}{2}du^{1-\frac{2}{d}}\Big) + u\Big],$$
$$\lim_{d\to 2} f_d(u) = \frac{1}{4}\Big(-1 - \ln u + u\Big).$$

The variation of $E_S + E_C$ with respect to r gives the familiar relation

$$E_S = 2E_C. \quad (1)$$

The bulk energy is given by

$$E_B = un'\Big[E_0 + \frac{K}{18}\Big(1 - \frac{n'}{n_s}\Big)^2\Big],$$

with E_0 and K being the binding energy per baryon and the incompressibility. The variation of $E_S + E_C + E_B$ with respect to n' gives an equation that determines u [1]. That for the cylindrical and spherical hole morphologies is similar. Thus, n', r, and r_c for each n and morphology are determined. The electron energy that is common to all morphologies are irrelevant to energy comparison but can be given by

$$E_e = \frac{3}{4}\hbar c\Big(3\pi^2 n_e\Big)^{\frac{1}{3}} n_e,$$
$$n_e = xn,$$

as an ultrarelativistic gas [20]. Adopting the parameter set $x = 0.3$, $E_0 = -11.4$ MeV, $K = 291$ MeV, $n_s = 0.147$ fm^{-3}, and $\sigma = 0.74$ MeV/fm^2 given in Ref. [1] and relevant to stellar collapses, the relative energy density and the cell size are obtained as in Figs. 1 and 2. Figure 1 indicates the sequential shape change as the density change. The cell sizes of the spherical nucleus and hole in Fig. 2 are used later.

In the above model for the five known morphologies, $E_S \propto r^{-1}$ and $E_C \propto r^2$ are given independently and accordingly the variation of their sum with respect to r leads to Eq. (1). In the case of the OBDD structure, however, E_S and E_C can not be represented simply (at least to the author's knowledge). Alternatively, we can utilize

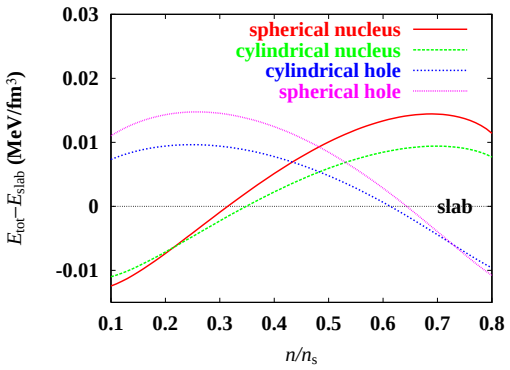

FIG. 1: (Color online) Total energy density of each morphology relative to that of the slab, as a function of the average density.

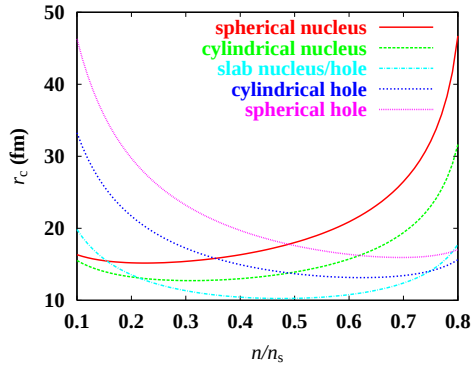

FIG. 2: (Color online) Cell size of each morphology as a function of the average density.

the non-dimensionalized area-volume relation given for a cubic cell in Ref. [17]. With a lattice parameter X, this gives S/X^2 as a function of V/X^3. Consequently, the surface energy density is given by

$$E_S = \sigma\frac{S}{X^2}\frac{1}{X},$$

as a function of $u \equiv V/X^3$. The relation between u and n is taken from the $d = 3$ (nucleus or hole) case. Assuming that Eq. (1) holds also for this morphology, the Coulomb energy is automatically determined. This means that the total energy can be obtained since E_B and E_e are independent of morphology. The area-volume relation in Fig. 3 for the OBDD morphology was adapted from Fig. 1(b) in Ref. [17] for the single-diamond structure. Since the relation

$$\frac{dS}{dV} = 2H$$

can be derived form the first variational formula of area, the nuclear OBDD ($V/X^3 < 0.5$) is a family of surfaces with $H > 0$, while the hole OBDD ($V/X^3 > 0.5$) is that with $H < 0$ (see Fig. 4 in Ref. [11]). Although the $H > 0$ and $H < 0$ parts are connected smoothly in the case

of the single-diamond structure, their curvatures are discontinuous in the case of the double-diamond structure. This indicates that the lamella structure exists between them.

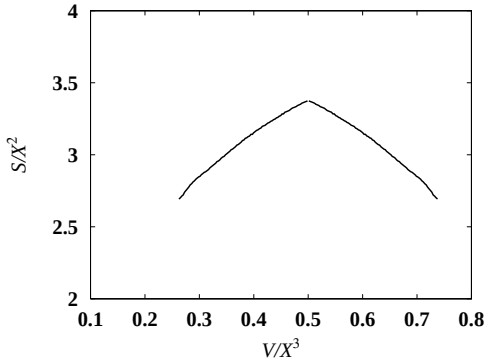

FIG. 3: Normalized area-volume relation for the OBDD structure. This is adapted from that for the single-diamond structure in Ref. [17]. The derivative is proportional to the mean curvature.

Assuming $X = \left(\frac{4\pi}{3}\right)^{1/3} r_c(d=3)$ (see Fig. 2), we estimate the energy density of the OBDD phase. The result is shown in Fig. 4. This figure indicates that this simple estimate gives 25 – 30 keV/fm^3 higher energy for the OBDD structure than the slab. Qualitatively, this result is consistent with that the OBDD phase is not the ground state at any composition in block copolymers. In the previous works, the "cross" phase in Ref. [4] and the "mixed" phase in Ref. [5] might correspond to the OBDD phase although our estimate gives higher energy. We did not try to change the parameters given in Ref. [1] because all parameters correlate and they require Skyrme model calculation that is beyond the scope of the present simple estimate.

To summarize, we proposed to identify the new "intermediate" morphology in subsaturation nuclear matter

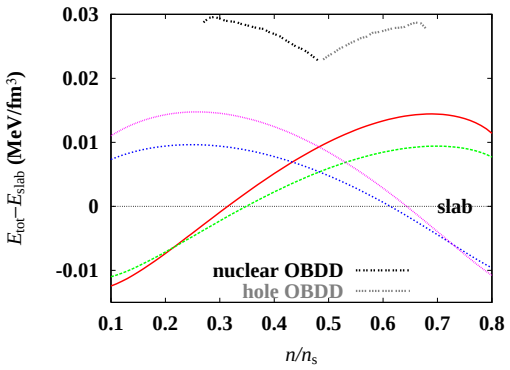

FIG. 4: (Color online) Total energy density of nuclear and hole OBDD structures relative to that of the slab, as functions of the average density. Those for the known morphologies are the same as in Fig. 1.

observed in a recent QMD simulation with the ordered bicontinuous double-diamond structure known in block copolymers. We estimated its energy density in a hybrid manner — incorporating the normalized area-volume relation given mathematically in a literature into the nuclear liquid drop model. The resulting energy density is higher than the other five known morphologies; this is qualitatively consistent with the results for block copolymers.

Note added in proof

After submission of the manuscript, the author found that another constant mean curvature surface, the bicontinuous double-gyroid structure similar to the OBDD but consisting of three-fold junctions [21], is favored in block copolymers. In the nuclear case, mathematical surfaces are meaningful as an idealization and therefore it would be difficult to distinguish them.

The author thanks S. Ei for directing his interest to block copolymers.

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